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This research program was concerned with the study of the phenomena of quantum superconductivity and with the evaluation of the potential for the application of such phenomena in superconducting electronic devices. The initial research efforts were focused on the study of the origin of 1/f noise in superconducting tunnel junctions and in ultra-high frequency effects and non-equilibrium phenomena in Josephson junctions. The 1/f noise research involved the study of the dynamic behavior of individual electronic defects in very small device structures. It led to the discovery of the reversible fluctuations of atomic defects between metastable configurations as the microscopic source of 1/f noise in tunnel junctions and in compound semiconductor device structures and to the discovery of strong, apparently lattice-mediated interactions, between these defects. With the discovery of high temperature superconductivity (HTS) partway through this research project a major portion of the research effort was redirected to the effort to fabricate and study high quality cuprate oxide superconductor thin films. This effort resulted in the first demonstration of the *in-situ* growth of HTS films and in their application in a number of studies that probed the nature of this new phenomenon.

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PRINCIPAL INVESTIGATOR: Robert A. Buhrman

CONTRACTOR: Cornell University

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INTRODUCTION

At the beginning of this research program, three related and complementary research projects in the area of quantum superconductivity were proposed and approved for funding. These were:

(1) An effort to examine the feasibility of a technologically competitive three-terminal device based on non-equilibrium superconductivity;

(2) Experiments to study the ultrahigh frequency behavior of Josephson tunnel junctions, with one objective being to develop a terahertz local oscillator for SIS mixer applications.

(3) A study of the origins and fundamental nature of 1/f noise in tunnel junctions and dc SQUIDs, and, by extension, in electronic systems in general.

The overall thrust of this research program, which was a continuation of previous ONR supported research efforts at Cornell in the area of quantum superconducting device research, was to address key research problems in the areas of quantum superconductivity that might lead either to enhanced quantum device performance or to new device applications, and to develop the materials science and nanofabrication technology necessary for the continued advancement of superconductive electronics.

Midway through the first two years of this research program, the experimental evidence was becoming clear that a non-equilibrium superconducting device would only be feasible, if at all, if a low carrier density superconductor was employed. This, and unresolved fundamental questions regarding the nature of the superconducting state in oxide materials, led to the initiation of an additional effort in the thin film growth, and study of superconductive electrodynamics, of the perovskite superconductor $Ba(Pb_{(1-x)}\text{-}Bi_x)O_3$. While our interest at that point in the perovskite superconductors was timely, we of course did not anticipate the astounding discoveries of the high temperature copper oxide superconductors that began to be announced in late 1986 and early 1987. These discoveries of course immediately affected all research in superconductivity, and it was quite obvious that the unique research opportunity and challenge that this development presented should be vigorously pursued. Consequently, the research focus of this research program shifted from the $Ba(Pb\text{-}Bi)O_3$ system, and from the completed investigation of non-equilibrium superconductivity devices, to the study of copper oxide superconductors. To support an expanded effort in this extremely important area, some additional funding was obtained for this research program from ONR to meet some acute capital equipment and staff needs. The overall focus of the research effort thus moved away from the quantum device development area, and moved towards the study of the basic physics and materials issues involved with understanding and ultimately manipulating the amazing phenomena of high temperature superconductivity.

At the same time we held that it would be inappropriate and wasteful to immediately abandon our three on-going efforts in low temperature superconductivity and cryogenic studies of individual electronic defect phenomena. These projects were important before the discovery of high temperature superconductivity and remained important after the discovery. Thus we established a program that had the greater emphasis on HTS research but which continued, at a somewhat reduced level, our prior activities in the first two project areas given above. This approach proved to be quite successful. As a result during this research program we completed our study and analysis of three-terminal superconducting devices[3] *(note the references are to the list of publications resulting from this research contract which is found at the end of this final report),* demonstrated the feasibility of a terahertz local oscillator that utilized the ac Josephson effect[8], completed a fundamental and unique study of the behavior of individual atomic scale defects in electronic systems[1,3,6], and developed a highly successful research capability in the area of high temperature superconducting thin films[10-13].

THREE-TERMINAL SUPERCONDUCTING DEVICE RESEARCH

During the previous ONR supported research program at Cornell and in the first two years of the program we were engaged in an effort to develop a successful three-terminal, transistor-like, device based on superconductivity. While we examined several alternative device concepts, the experimental focus of the effort was on the use of non-equilibrium, gas-suppression, effects to produce the desired response in the shared electrode of a double tunnel junction structure. Our basic device was similar in concept to the widely publicized QUITERON of Faris and coworkers as well as to the CLINK device of Langenberg and coworkers which was first studied over ten years ago. Our particular distinction was that we were using submicron-fabrication techniques to produce a device structure that might have some hope of attaining a respectable switching speed. The experiments were also designed to establish as clearly as possible the basic feasibility of such a device. Unfortunately, our conclusion from these experiments was that, with one possible exception, non-equilibrium superconductivity holds little promise for the development of a successful three-terminal device, barring the discovery of some new and unusual physics in what is a very well-studied area. A paper(3) summarizing our results in this area was published in 1986 and this research effort was completed.

The one possible exception that was mentioned above is dependent upon the successful synthesis of very low carrier density superconducting films. An example of such a material was $Ba(Pb_{(1-x)}-Bi_x)O_3$ oxide which was under very active study in Japan. If the number of free carriers n_e in a superconductor is sufficiently below that of typical metals, then it should be much easier to perturb the system with electron injection. Our studies indicated that a factor of ten reduction in n_e could yield a competitive non-equilibrium device. But it was apparent that there were basic materials and physics issues regarding this exotic material that needed to be addressed first before quantum device applications could be seriously evaluated. Consequently we began a research effort focused on the first step of fabrication high quality $Ba(Pb_{(1-x)}-Bi_x)O_3$ thin films. As indicated in the introduction the discovery of high temperature superconductivity almost at the same time quickly led to a postponement of this research effort in favor of a strong effort focused on the cuprate superconductors.

THE ULTRA-HIGH FREQUENCY LIMITS OF THE AC JOSEPHSON EFFECT

At the time of the beginning of this research program, many years of research into quantum superconductivity had clearly demonstrated a superior capability for the low-noise detection of mixing of millimeter and submillimeter wave radiation. Two important objectives of research in this area were then to extend SIS mixer performance further up into the terahertz region and to construct integrated mixer-local oscillator-amplifier receiver systems. But if SIS mixers and Josephson local oscillators were to be extended to terahertz frequencies, it was necessary that high critical current density, submicron junctions be fabricated and that the behavior of tunnel junctions close to and above the gap frequency be well understood.

For the past several years we had been engaged in efforts to produce reliable, high critical current density, low capacitance tunnel junctions comparable of responding effectively at ultra-high frequencies and to study their ac Josephson behavior at such frequencies. The reactive ion beam oxidation (RIBO) technique which we developed some time ago proved to be very well suited to this task of producing very thin, ≤ 1 nm, tunnel barriers. In the mixed refractor metal-soft metal tunnel junction systems we were able to routinely produce junctions with $J_c > 10^5$ A/cm^2, sufficient to extend the cutoff frequency of such junctions to well above 1 THz.

By producing coupled pairs of such junctions we were able to examine the ultrahigh frequency nature of the ac Josephson effect by using one junction as a local oscillator and the other as a radiation detector(8). This work demonstrated that quite adequate amounts of local oscillator radiation could be coupled to a superconducting mixer junction in the terahertz region. On the more fundamental side it has also demonstrated that the current theoretical understanding of actual Josephson junctions at and above the gap frequency is not in close accord with experiment. The cause of this disagreement remains unresolved but is most likely attributable to additional scattering mechanism in the superconducting electrodes which are not considered in the standard theory of the Josephson junction.

While this project was quite successful, as a result of consultation with the ONR program officer, it was decided that it would bee more appropriate that a continuation of the effort would be made as a part of the Terahertz Technology program of the Air Force's SDIO-IST program. Consequently the ONR support for this project terminated in the end of 1986.

INDIVIDUAL ATOMIC DEFECT TUNNELING SPECTROSCOPY

The originally proposed research on the studies of individual defect states in Josephson tunnel junctions and in semiconductor microstructures continued to a successful conclusion at the end of this research contract. This research grew out of our earlier discovery that the charging and discharging of single slow electron trap states could be observed directly through measurement of the resulting discrete two-level resistance fluctuations of very small tunnel junctions. The detailed behavior of these trap states involved the microscopic displacement of defect atoms as part of the filling and emptying process. It is this atomic motion that makes the process sufficiently slow that it can be observed experimentally. We determined that these slow trap states are the source of 1/f noise in Josephson tunnel structures and characterized the behavior of different tunnel barriers(1,2,4,7). Through careful study of the dynamics of these individual trap states we then determined that there are strong interactions between trap states that can lead to quite complex behavior, with, for example, the occupancy of one state promoting the occupancy of another nearby state(1,7). Moreover, we established that such interactions occur not only in superconducting tunnel junctions but also in GaAs-AlGaAs-GaAs tunneling devices and in MOS tunnel diodes. Such trap-trap interactions have never been reported previously. We also were able to determine that the effect of an applied tunnel current on the trap state is to speed up the switching of the state as a result of the process of inelastic scattering of the tunneling electrons by the defect.

These effects were observed both in MIM and GaAs micrometer scale devices. Although the details differed in each system studied, the general ehavior appears universal. These studies into the dynamics and interactions of individual electronic defects in insulating and semiconducting systems broke new ground in an area that is of both fundamental and applied interest. We consider this work, which has been widely cited and which has led to a much more basic understanding of defects and 1/f noise is electronic systems, to have been a major success of this ONR supported research program.

HIGH TEMPERATURE SUPERCONDUCTIVITY RESEARCH

As indicated, after the discovery of high temperature superconductivity, midway through this research period, the greater portion of this ONR supported research program became focused on high temperature superconductivity. This effort had as a major objective the development of techniques for the successful production of HTS materials, chiefly thin films, of a sufficiently high quality, and in a manner, that would then permit detailed study of the basic physics and materials science of these superconducting systems. Apart from a collaborative effort(5,6,9) with Professor Al Sievers' group in the Physics Department at Cornell on the far infrared properties of the new superconductors in bulk polycrystals and single crystals, this effort initially concentrated on developing a lower temperature, in-situ process for the growth of well oriented, high critical-current-density thin films of the 123 material. We were the first group, at least in the United States, to successfully demonstrate an in-situ process(10,11), and we continually worked to improve this process over the final months of the research contract. At the end of that time we were successfully growing epitaxial single crystal c-axis normal films on MgO substrates with a maximum substrate temperature of 600-660 C. Our best critical current density result on such films was 5 x 10^6 A/cm^2 at 4.2 K as measured by direct transport on 10 μm wide microstrips. Our reactive evaporation process could be used at still lower temperatures, but the quality of the superconducting transition and film orientation were found to deteriorate for a growth temperature below 600 C. As indicated above, our emphasis was on obtaining good quality material for research and thus we did not focus on continually lowering the growth temperature simply to set a temporary record but aimed at developing processes that allowed us to address basic science and materials issues in HTS.

We employed the films produced in these growth experiments in a number of investigations, both in our laboratory and in collaboration with several other groups. These included detailed high resolution electron microscopy studies of the thin film material[13], rf surface loss and dc transport measurements[11], far infrared optical studies of the energy gap and related electrodynamics[9], and the first picosecond pulse propagation studies of HTS transmission lines[12]. Again, our basic program objective was to develop a detailed understanding of HTS thin films and of what the fundamental electrodynamic behavior of this class of superconductors is.

As part of transport and transmission line results we developed successful photolithography techniques for the patterning of HTS thin films to micrometer and submicrometer dimensions. This resulted in no discernible deterioration of the superconductive properties of the film. We were the first to have accomplished this, in part because the smooth, clean nature of our in-situ formed films were far more suited for high resolution etching and processing than were the much rougher, large grain size films that are obtained from deposition processes that required an extended high temperature anneal.

Summary of Major Research Highlights During Contract Period

We consider the major research accomplishments during this research program to have been:

1. The discovery and unique identification of the reversible fluctuations of atomic defects between metastable configurations as the microscopic source of 1/f noise in MIM and compound semiconductor device structures and the observation of strong, apparently lattice-mediated interactions, between these defects.
2. The characterization of the different levels of 1/f noise and hence of localized electron states in the different types of Josephson tunnel junctions. From the results of this work the type of junction can be selected that is best for use in low noise SQUID applications.
3. The completion of a detailed study of the ac Josephson effect at the gap frequency. We demonstrated the feasibility of using small area, high critical current density tunnel junctions as terahertz local oscillators while also establishing that the approximate solutions of the Werthamer tunnel junction equations do not successfully describe the ultrahigh frequency behavior of these junctions.
4. The development and demonstration of the first successful technique for the in-situ formation, in the deposition chamber, and during the deposition process, of the superconducting 1-2-3 phase of yttrium-barium-copper-oxide thin films.
5. The first successful patterning of 123 thin films to micron and sub-micron dimensions with no detectable deterioration of the superconductive properties of the patterned film and the development of a successful technique for the formation of very low resistance, ohmic contacts to the films.
6. The successful epitaxial growth of smooth, high quality 123 thin films on MgO single crystal substrates both by reactive evaporation and by reactive sputtering with very clean interfaces, as revealed by cross-section transmission electron microscopy, between substrate and film.
7. The first application of HTS superconducting transmission lines in picosecond electrical pulse propagation experiments.

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